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PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of

Masahiro FURUKAWA et al.

Attn: PCT Branch

Application No. New U.S. National Stage of PCT/JP03/14726

Filed: May 17, 2005

Docket No.: 123927

For: SILICON CARBIDE POROUS BODY, PROCESS FOR PRODUCING THE
SAME AND HONEYCOMB STRUCTURE

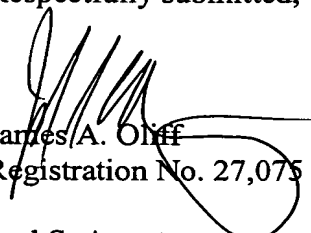
TRANSLATION OF THE ANNEXES TO THE
INTERNATIONAL PRELIMINARY EXAMINATION REPORT

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

Attached hereto is a translation of the annexes to the International Preliminary Examination Report (Form PCT/IPEA/409). The attached translated material replaces the material in the specification at pages 5-9, 12, 15, 20, 21, 29, and claims 1-14.

Respectfully submitted,


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Translation

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PATENT COOPERATION TREATY

PCT/JP2003/014726



PCT

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY
(Chapter II of the Patent Cooperation Treaty)

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference WA-0866	FOR FURTHER ACTION See Form PCT/IPEA/416	
International application No. PCT/JP2003/014726	International filing date (day/month/year) 19 November 2003 (19.11.2003)	Priority date (day/month/year) 20 November 2002 (20.11.2002)
International Patent Classification (IPC) or national classification and IPC C04B 35/576, 38/00		
Applicant NGK INSULATORS, LTD.		

- This report is the international preliminary examination report, established by this International Preliminary Examining Authority under Article 35 and transmitted to the applicant according to Article 36.
- This REPORT consists of a total of 3 sheets, including this cover sheet.
- This report is also accompanied by ANNEXES, comprising:
 - ☒ (sent to the applicant and to the International Bureau) a total of 11 sheets, as follows:
 - ☒ sheets of the description, claims and/or drawings which have been amended and are the basis of this report and/or sheets containing rectifications authorized by this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions).
 - ☐ sheets which supersede earlier sheets, but which this Authority considers contain an amendment that goes beyond the disclosure in the international application as filed, as indicated in item 4 of Box No. I and the Supplemental Box.
 - ☐ (sent to the International Bureau only) a total of (indicate type and number of electronic carrier(s)) _____, containing a sequence listing and/or tables related thereto, in computer readable form only, as indicated in the Supplemental Box Relating to Sequence Listing (see Section 802 of the Administrative Instructions).

- This report contains indications relating to the following items:
 - ☒ Box No. I Basis of the report
 - ☐ Box No. II Priority
 - ☐ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
 - ☐ Box No. IV Lack of unity of invention
 - ☒ Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
 - ☐ Box No. VI Certain documents cited
 - ☐ Box No. VII Certain defects in the international application
 - ☐ Box No. VIII Certain observations on the international application

Date of submission of the demand 09 April 2004 (09.04.2004)	Date of completion of this report 09 December 2004 (09.12.2004)
Name and mailing address of the IPEA/JP	Authorized officer
Facsimile No.	Telephone No.

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No.

PCT/JP2003/014726

Box No. I Basis of the report

1. With regard to the language, this report is based on the international application in the language in which it was filed, unless otherwise indicated under this item.

- ☐ This report is based on translations from the original language into the following language _____, which is language of a translation furnished for the purpose of:
- ☐ international search (under Rules 12.3 and 23.1(b))
- ☐ publication of the international application (under Rule 12.4)
- ☐ international preliminary examination (under Rules 55.2 and/or 55.3)

2. With regard to the elements of the international application, this report is based on *(replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report)*:

- ☐ The international application as originally filed/furnished
- ☒ the description:
- pages _____ 1, 2, 6, 9, 10, 13-15, 17-21 _____, as originally filed/furnished
- pages* _____ 7, 16 _____ received by this Authority on _____ 04 August 2004 (04.08.2004)
- pages* _____ 3-5, 8, 11, 12 _____ received by this Authority on _____ 29 November 2004 (29.11.2004)
- ☒ the claims:
- pages _____ 5, 10 _____, as originally filed/furnished
- pages* _____, as amended (together with any statement) under Article 19
- pages* _____ 1, 3, 4, 7-9, 11, 12, _____ received by this Authority on _____ 04 August 2004 (04.08.2004)
- pages* _____ received by this Authority on _____
- ☒ the drawings:
- pages _____ 1-4 _____, as originally filed/furnished
- pages* _____ received by this Authority on _____
- pages* _____ received by this Authority on _____
- ☐ a sequence listing and/or any related table(s) – see Supplemental Box Relating to Sequence Listing.

3. ☒ The amendments have resulted in the cancellation of:

- ☐ the description, pages _____
- ☒ the claims, Nos. _____ 2, 6, 13 _____
- ☐ the drawings, sheets/figs _____
- ☐ the sequence listing (*specify*): _____
- ☐ any table(s) related to sequence listing (*specify*): _____

4. ☐ This report has been established as if (some of) the amendments annexed to this report and listed below had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).

- ☐ the description, pages _____
- ☐ the claims, Nos. _____
- ☐ the drawings, sheets/figs _____
- ☐ the sequence listing (*specify*): _____
- ☐ any table(s) related to sequence listing (*specify*): _____

* If item 4 applies, some or all of those sheets may be marked "superseded."

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No.

PCT/JP03/14726

Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**1. Statement**

Novelty (N)	Claims	1, 3-5, 7-12, 14	YES
	Claims		NO
Inventive step (IS)	Claims	1, 3-5, 7-12, 14	YES
	Claims		NO
Industrial applicability (IA)	Claims	1, 3-5, 7-12, 14	YES
	Claims		NO

2. Citations and explanations (Rule 70.7)

Document 1: WO, 02/81406, A1 (NGK Insulators, Ltd.)

Document 1 cited in the ISR describes that a silicon carbide-based porous body comprising silicon carbide particles and metallic silicon has an amorphous or crystalline silicic acid salt compound phase and that the silicon carbide particles are bonded to each other with the metallic silicon and/or the silicic acid salt compound phase. Further, an embodiment described therein relates to cordierite, which is a Mg-Al-Si crystalline oxide as a specific example of the silicic acid salt compound phase.

However, document 1 neither describes nor suggests that a structure with embedded fine porous sections is obtained when the silicic acid salt compound is a Sr-Al-Si amorphous oxide.

Therefore, the invention of the subject application (claims 1, 3-5, 7-12, 14) appears to possess novelty and involve an inventive step.

[1] A silicon carbide porous body comprising silicon carbide particles which are aggregates and metallic silicon which is a bonding material, bonded together in such a manner that pores are retained between the silicon carbide particles and/or between the silicon carbide particle and metallic silicon, characterized in that an oxide phase containing oxides of silicon, aluminum, and alkaline earth metal is buried in at least some of fine pore portions having a minimum distance of 10 μm or less between the surfaces of the silicon carbide particles or between the surfaces of the silicon carbide particle and metallic silicon among the pores, and a ratio of a total volume (pore volume of the fine pore portion) of portions in which the oxide phase is not buried among the fine pore portions is 20% or less with respect to a total volume (total pore volume) of portions in which the oxide phase is not buried among the pores including the fine pore portions.

[2] The silicon carbide porous body according to the above [1], wherein the alkaline earth metal is strontium.

[3] The silicon carbide porous body according to the above [1] or [2], wherein a plane image obtained by photographing a cut face of the silicon carbide porous body cut with a predetermined plane is subjected to an image analysis process, and divided into a specified pore portion originating from the portion in which the oxide phase is not buried in the pore including the fine pore portion, a

specified silicon carbide particle portion originating from the silicon carbide particle, a specified metallic silicon portion originating from metallic silicon, and a specified oxide phase portion originating from the oxide phase, and a relation of the following equation (1) is satisfied by a total length (contact length) L (mm/mm²) per unit area (1 mm²) of a portion with which the silicon carbide particle portion, the metallic silicon portion, and the oxide phase portion are brought into contact on the divided plane image, and a porosity ϵ (%) of the silicon carbide porous body:

$$L \leq -1.0\epsilon + 90 \dots (1).$$

[4] The silicon carbide porous body according to any one of the above [1] to [3], wherein a ratio of a total area of portions with which the silicon carbide particle and the oxide phase are brought into contact is in a range of 10 to 70% with respect to a total area of portions with which the silicon carbide particle, metallic silicon, and oxide phase are brought into contact.

[5] The silicon carbide porous body according to the above [4], wherein the ratio of the total area of the portions with which the silicon carbide particle and the oxide phase are brought into contact is in a range of 25 to 50% with respect to the total area of the portions with which the silicon carbide particle, metallic silicon, and oxide phase are brought into contact.

[6] The silicon carbide porous body according to any one of the above [2] to [5], wherein the oxide phase is

amorphous, the oxide phase contains all oxides (SrO , Al_2O_3 , SiO_2) of strontium, aluminum, and silicon, and a content ratio ($\text{SrO}:\text{Al}_2\text{O}_3:\text{SiO}_2$) of the respective oxides of strontium, aluminum, and silicon in the oxide phase is in a range of
5 (1.0:0.1:1.0) to (1.0:1.0:3.0) in accordance with each substance amount ratio (molar ratio).

[7] The silicon carbide porous body according to the above [6], wherein melting temperatures of the oxides (SrO , Al_2O_3 , SiO_2) are in a range of 1000 to 1400°C.

10 [8] The silicon carbide porous body according to the above [6] or [7], wherein melting viscosity of the oxide phase is lower than that of metallic silicon.

[9] The silicon carbide porous body according to any one of the above [6] to [8], wherein a ratio of mass of
15 the oxide phase is in a range of 1.0 to 10.0 mass% with respect to a total mass of the silicon carbide particle and metallic silicon.

[10] The silicon carbide porous body according to the above [9], wherein a ratio of mass of the oxide phase
20 is in a range of 4.0 to 8.0 mass% with respect to a total mass of the silicon carbide particle and metallic silicon.

[11] A honeycomb structure comprising: the silicon carbide porous body according to any one of the above [1] to [10].

25 [12] A process for producing a silicon carbide porous body, characterized by: adding, to silicon carbide particles and metallic silicon, compound containing

strontium, aluminum, and silicon in a range of 1.0 to 10.0 parts by mass in terms of oxides (SrO , Al_2O_3 , SiO_2) with respect to a total of 100 parts by mass of the silicon carbide particles and metallic silicon to obtain a raw

5 material; forming the obtained raw material into a predetermined shape to obtain a formed article; degreasing and thereafter firing the obtained formed article; and burying an oxide phase containing the respective oxides of silicon, aluminum, and alkaline earth metal in at least

10 some of fine pore portions having a minimum distance of 10 μm or less between the surfaces of the respective silicon carbide particles or between the surfaces of the silicon carbide particle and metallic silicon among the pores formed between the silicon carbide particles in such a

15 manner that a ratio of a total volume (pore volume of the fine pore portion) of portions in which the oxide phase is not buried among the fine pore portions is 20% or less with respect to a total volume (total pore volume) of portions in which the oxide phase is not buried among the pores

20 including the fine pore portions to obtain the porous body having a porous structure.

[13] The process for producing the silicon carbide porous body according to claim 12, wherein a type and/or an adding amount of the compound containing strontium,

25 aluminum, and silicon are adjusted in such a manner that a content ratio ($\text{SrO}:\text{Al}_2\text{O}_3:\text{SiO}_2$) of the oxides of strontium, aluminum, and silicon is in a range of (1.0:0.1:1.0) to

(1.0:1.0:3.0) in each substance amount ratio (molar ratio), the oxides being contained in the oxide phase constituting the porous body having the porous structure obtained by the firing.

5 [14] The process for producing the silicon carbide porous body according to the above [12] or [13], wherein an amount of the compound to be added to the silicon carbide particles and metallic silicon and containing strontium, aluminum, and silicon, converted into the respective oxides
10 (SrO , Al_2O_3 , SiO_2), is set to a range of 4.0 to 8.0 parts by mass with respect to a total amount of 100 parts by mass of the silicon carbide particles and metallic silicon.

Brief Description of the Drawings

15 FIG. 1 is a sectional view schematically showing one embodiment of a silicon carbide porous body of the present invention;

 FIG. 2 is a graph in which a contact length L (mm/mm^2) is plotted with respect to a value of a porosity ϵ (%) in the silicon carbide porous body of an example of the
20 present invention;

 FIG. 3 is an electron microscope photograph of a silicon carbide porous body of Example 1 of the present invention; and

25 FIG. 4 is an electron microscope photograph of a silicon carbide porous body of Example 2 of the present invention.

carbide particles 2, or between the surfaces of the silicon carbide particle 2 and metallic silicon 3 are the fine pore portions 6. In the present embodiment, the oxide phases 5 are buried at least some of the fine pore portions 6. It is to be noted that the oxide phase 5 may be buried in such a manner as to seal all the fine pore portions 6.

A volume of the portion in which any oxide phase 5 is not buried in the pore 4 including the above-described fine pore portion 6, and that of the portion in which any oxide phase 5 is buried in the fine pore portion 6 can be calculated from a pore diameter distribution measured, for example, using a mercury porosimeter or the like. The section of the silicon carbide porous body 1 is photographed by a scanning electron microscope (SEM) or the like, and images photographed in a plurality of sections are analyzed. The volumes may be calculated as integrated values.

Moreover, in the present embodiment, in a case where a total of lengths of portions with which the silicon carbide particle 2, metallic silicon 3, and oxide phase 5 are brought into contact per unit area (1 mm^2) is "contact length L (mm/mm^2)", porosity ϵ (%) of the silicon carbide porous body 1, and the above-described contact length L (mm/mm^2) preferably satisfy a relation of the following equation (1):

$$L \leq -1.0\epsilon + 90 \dots (1).$$

The above-described equation (1) is an evaluation

plurality of sections in a thickness direction are subjected to computer image analysis. A length (interface length) of the boundary line is extracted using an image analysis method similar to an evaluation method in which bonding strength is judged between the silicon carbide particles 2 in the above-described predetermined porosity, and the length is approximately usable. It is to be noted that from a viewpoint that the silicon carbide porous body 1 be obtained having high thermal conductivity and mechanical strength, the ratio of the total area of the portions with which the silicon carbide particle 2 and the oxide phase 5 are brought into contact is preferably 25 to 50% with respect to the total area of the portions with which the silicon carbide particle 2, metallic silicon 3, and oxide phase 5 are brought into contact.

In the present embodiment, the above-described alkaline earth metal is preferably strontium. Furthermore, preferably the oxide phase 5 is amorphous, the oxide phase 5 contains all oxides (SrO , Al_2O_3 , SiO_2) of strontium, aluminum, and silicon, and a content ratio ($\text{SrO}:\text{Al}_2\text{O}_3:\text{SiO}_2$) of the respective oxides of strontium, aluminum, and silicon in the oxide phase 5 is (1.0:0.1:1.0) to (1.0:1.0:3.0) in accordance with each substance amount ratio (molar ratio).

By this constitution, the respective oxides are regarded as a ternary compound system, and an eutectic point is lowered. Accordingly, during firing, an oxide

added in such a manner that the finally formed oxide phase contains at least one type of the alkaline earth metals, aluminum, and silicon. When a plurality of types is added, the added amounts may be mutually different or equal. In this case, a forming auxiliary agent such as an organic binder may be added if necessary. It is to be noted that the silicon carbide particle or metallic silicon sometimes contains a micro amount of impurities such as iron, aluminum, and calcium, but may be simply used, or may be subjected to chemical treatment such as chemical cleaning and refined for use. When an amount of compound to be added is less than 1.0 part by mass in terms of the respective oxides (SrO , Al_2O_3 , SiO_2), the strength of the obtained silicon carbide porous body cannot be sufficiently effectively enhanced. When the amount exceeds 10.0 parts by mass, the amount of the oxide phase formed by the compound is excessively large, therefore the firing contraction increases, and the porosity of the obtained silicon carbide porous body drops. For example, when the porous body is used as the filter like the DPF, the pressure loss excessively increases. It is preferable that strontium in the compound is contained in the form of strontium oxide (SrO) or strontium carbonate (SrCO_3) because the oxide phase can be efficiently formed, and the substance is easily obtainable and easily handled. Similarly, aluminum is preferably contained in the form of aluminum oxide (Al_2O_3) or metallic aluminum. It is to be

noted that, in this case, metallic aluminum may be contained as impurities of metallic silicon. Similarly, silicon is preferably contained in the form of silicon dioxide (SiO_2) or colloidal silica. It is to be noted that, in this case, silicon dioxide may be contained as an oxide film (SiO_2) with which the surface of the silicon carbide particle and/or metallic silicon is to be coated.

Next, the raw materials obtained in this manner are mixed and kneaded to constitute a clay for forming, this clay is formed into a predetermined shape such as a honeycomb shape, this clay is calcined, and the organic binder is degreased to obtain a formed article.

Next, the obtained formed article is fired, and an oxide phase containing the respective oxides of silicon, aluminum, and alkaline earth metal is buried in at least some of pores formed between the silicon carbide particles in such a manner that a ratio of a total volume (pore volume of the fine pore portion) of portions in which the oxide phase is not buried among the fine pore portions is 20% or less with respect to a total volume (total pore volume) of portions in which the oxide phase is not buried among the pores including the fine pore portions to obtain the porous body having a porous structure.

In the producing process of the present embodiment, a type and/or an adding amount of the compound containing strontium, aluminum, and silicon are preferably adjusted in such a manner that a content ratio ($\text{SrO}:\text{Al}_2\text{O}_3:\text{SiO}_2$) of the

flat face, the cut face may be appropriately polished. A plane image obtained by photographing the cut face using the scanning electron microscope (SEM) or the like was taken into a calculator including a personal computer (PC) using image take-in means such as a scanner. By a predetermined image analyzing method, the taken-in plane image was divided and extracted as the silicon carbide particle 2, metallic silicon 3, oxide phase 5, and portion (pore portion) in which any oxide phase 5 was not buried in the pore 4 including the fine pore portion 6 as shown in FIG. 1. When a predetermined image processing method was applied to a boundary among the extracted silicon carbide particle 2, metallic silicon 3, and oxide phase 5, a boundary line having a width for one pixel was extracted, and the total of the lengths per unit area (1 mm^2) was calculated as the contact length L (mm/mm^2). Obtained results are shown in Table 1. A graph in which the contact length L (mm/mm^2) is plotted with respect to the value of the porosity ϵ (%) is shown in FIG. 2. It is to be noted that a straight line in FIG. 2 was drawn based on a lower-limit value of the following equation (1):

$$L \leq -1.0\epsilon + 90 \dots (1).$$

Moreover, a ratio (%) (hereinafter sometimes referred to as the oxide bonding ratio (%)) of a total area of portions with which the silicon carbide particles and oxide phase were brought into contact was calculated with respect to a total area of portions with which the silicon

CLAIMS

1. A silicon carbide porous body comprising silicon carbide particles which are aggregates and metallic silicon which is a bonding material, bonded together in such a manner that pores are retained between the silicon carbide particles and/or between the silicon carbide particle and metallic silicon,

characterized in that an oxide phase containing oxides of silicon, aluminum, and alkaline earth metal is buried in at least some of fine pore portions having a minimum distance of 10 μm or less between the surfaces of the silicon carbide particles or between the surfaces of the silicon carbide particle and metallic silicon among the pores, and

a ratio of a total volume (pore volume of the fine pore portion) of portions in which the oxide phase is not buried among the fine pore portions is 20% or less with respect to a total volume (total pore volume) of portions in which the oxide phase is not buried among the pores including the fine pore portions.

2. The silicon carbide porous body according to claim 1, wherein the alkaline earth metal is strontium.

3. The silicon carbide porous body according to claim 1 or 2, wherein a plane image obtained by photographing a cut face of the silicon carbide porous body cut with a predetermined plane is subjected to an image analysis process, and divided into a specified pore portion

originating from the portion in which the oxide phase is not buried in the pore including the fine pore portion, a specified silicon carbide particle portion originating from the silicon carbide particle, a specified metallic silicon portion originating from metallic silicon, and a specified oxide phase portion originating from the oxide phase, and a relation of the following equation (1) is satisfied by a total length (contact length) L (mm/mm²) per unit area (1 mm²) of a portion with which the silicon carbide particle portion, the metallic silicon portion, and the oxide phase portion are brought into contact on the divided plane image, and a porosity ϵ (%) of the silicon carbide porous body:

$$L \leq -1.0\epsilon + 90 \dots (1).$$

4. The silicon carbide porous body according to any one of claims 1 to 3, wherein a ratio of a total area of portions with which the silicon carbide particle and the oxide phase are brought into contact is in a range of 10 to 70% with respect to a total area of portions with which the silicon carbide particle, metallic silicon, and oxide phase are brought into contact.

5. The silicon carbide porous body according to claim 4, wherein the ratio of the total area of the portions with which the silicon carbide particle and the oxide phase are brought into contact is in a range of 25 to 50% with respect to the total area of the portions with which the silicon carbide particle, metallic silicon, and oxide phase are brought into contact.

6. The silicon carbide porous body according to any one of claims 2 to 5, wherein the oxide phase is amorphous, the oxide phase contains all oxides (SrO , Al_2O_3 , SiO_2) of strontium, aluminum, and silicon, and a content ratio
5 ($\text{SrO}:\text{Al}_2\text{O}_3:\text{SiO}_2$) of the respective oxides of strontium, aluminum, and silicon in the oxide phase is in a range of (1.0:0.1:1.0) to (1.0:1.0:3.0) in accordance with each substance amount ratio (molar ratio).

7. The silicon carbide porous body according to claim 6,
10 wherein melting temperatures of the oxides (SrO , Al_2O_3 , SiO_2) are in a range of 1000 to 1400°C.

8. The silicon carbide porous body according to claim 6 or 7, wherein melting viscosity of the oxide phase is lower than that of metallic silicon.

15 9. The silicon carbide porous body according to any one of claims 6 to 8, wherein a ratio of mass of the oxide phase is in a range of 1.0 to 10.0 mass% with respect to a total mass of the silicon carbide particle and metallic silicon.

20 10. The silicon carbide porous body according to claim 9, wherein a ratio of mass of the oxide phase is in a range of 4.0 to 8.0 mass% with respect to a total mass of the silicon carbide particle and metallic silicon.

11. A honeycomb structure comprising: the silicon
25 carbide porous body according to any one of claims 1 to 10.

12. A process for producing a silicon carbide porous body, characterized by: adding, to silicon carbide

particles and metallic silicon, compound containing strontium, aluminum, and silicon in a range of 1.0 to 10.0 parts by mass in terms of oxides (SrO , Al_2O_3 , SiO_2) with respect to a total of 100 parts by mass of the silicon carbide particles and metallic silicon to obtain a raw material; forming the obtained raw material into a predetermined shape to obtain a formed article; degreasing and thereafter firing the obtained formed article; and burying an oxide phase containing the respective oxides of silicon, aluminum, and alkaline earth metal in at least some of fine pore portion having a minimum distance of 10 μm or less between the surfaces of the respective silicon carbide particles or between the surfaces of the silicon carbide particle and metallic silicon among the pores formed between the silicon carbide particles in such a manner that a ratio of a total volume (pore volume of the fine pore portion) of portions in which the oxide phase is not buried among the fine pore portions is 20% or less with respect to a total volume (total pore volume) of portions in which the oxide phase is not buried among the pores including the fine pore portions to obtain the porous body having a porous structure.

13. The process for producing the silicon carbide porous body according to claim 12, wherein a type and/or an adding amount of the compound containing strontium, aluminum, and silicon are adjusted in such a manner that a content ratio ($\text{SrO}:\text{Al}_2\text{O}_3:\text{SiO}_2$) of the oxides of strontium, aluminum, and

silicon is in a range of (1.0:0.1:1.0) to (1.0:1.0:3.0) in each substance amount ratio (molar ratio), the oxides being contained in the oxide phase constituting the porous body having the porous structure obtained by the firing.

- 5 14. The process for producing the silicon carbide porous body according to claims 12 and 13, wherein an amount of the compound to be added to the silicon carbide particles and metallic silicon and containing strontium, aluminum, and silicon, converted into the respective oxides (SrO ,
10 Al_2O_3 , SiO_2), is set to a range of 4.0 to 8.0 parts by mass with respect to a total amount of 100 parts by mass of the silicon carbide particles and metallic silicon.